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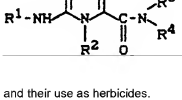
(71) Applicant: **BAYER AG**

D-5090 Leverkusen 1 Bayerwerk (DE)

(72) Inventor: **Findeisen, Kurt, Dr.**
Dünfelder Strasse 28
D-5090 Leverkusen 1 (DE)
Inventor: **Santel, Hans-Joachim, Dr.**
Grünstrasse 9a
D-5090 Leverkusen 1 (DE)
Inventor: **Lürssen, Klaus, Dr.**
August-Kierspel-Strasse 145
D-5060 Bergisch Gladbach 2 (DE)
Inventor: **Schmidt, Robert R., Dr.**
Im Waldwinkel 110
D-5060 Bergisch Gladbach 2 (DE)

(54) **3-amino-5-aminocarbonyl-1,2,4-triazole derivatives.**

(57) Novel 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of the general formula (I),



(I)

several methods for their preparation, and their use as herbicides.

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3-AMINO-5-AMINOCARBONYL-1,2,4-TRIAZOLE DERIVATIVES

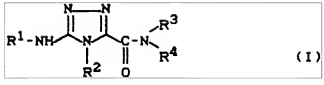
The invention relates to novel 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives, several methods for their preparation, and their use as herbicides.

It is known that certain nitrogen heterocycles (cf., for example, K. H. Büchel "Pflanzenschutz und Schädlingsbekämpfung" [Plant Protection and Pest Control] p. 170, Thieme Verlag Stuttgart, 1977) have herbicidal properties.

In addition, certain substituted triazoles are the subject matter of one of our own antecedent but not previously published patent applications (cf. DE-P 38 09 053 of March 18, 1988).

However, the herbicidal activity of known compounds is not entirely satisfactory in all application areas.

Novel 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of the general formula (I) have now been found,



(I)

in which

R^1 represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl or halogenoalkinyl having in each case 2 to 8 carbon atoms and 1 to 15, or 13 respectively, identical or different halogen atoms, alkoxyalkyl having 1 to 6 carbon atoms in the individual alkyl moieties, or represents cycloalkyl having 3 to 7 carbon atoms, or represents cycloalkylalkyl having 3 to 7 carbon atoms in the cycloalkyl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, or represents aralkyl which has 6 to 10 carbon atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 3 hetero atoms, in particular, nitrogen, oxygen and/or sulfur, whereby in each case suitable substituents are: halogen, cyano, nitro and in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy or halogenoalkylthio, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms,

R^2 represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl having 2 to 8 carbon atoms and 1 to 15 identical or different halogen atoms, halogenoalkinyl having 2 to 8 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxyalkyl having 1 to 6 carbon atoms in each of the individual alkyl moieties, or represents cycloalkylalkyl or cycloalkyl, in each case having 3 to 7 carbon atoms in the cycloalkyl moiety and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, or represents aralkyl or aryl, each of which has 6 to 10 carbon atoms in the aryl moiety and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable aryl substituents are: halogen, cyano, nitro and in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy or halogenoalkylthio, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms, and

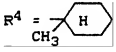
R^3 and R^4 independently of one another in each case represent hydrogen, or in each case represent straight-chain or branched alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl or halogenoalkinyl, in each case having 2 to 8 carbon atoms and 1 to 15, or 13 respectively, identical or different halogen atoms, cyanoalkyl having 1 to 8 carbon atoms, hydroxyalkyl having 1 to 8 carbon atoms and 1 to 6 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, in each case having up to 6 carbon atoms in the individual alkyl or alkenyl moieties, alkylaminoalkyl or dialkylaminoalkyl, in each case having 1 to 6 carbon atoms in

the individual alkyl moieties, or cycloalkyl, cycloalkylalkyl, cycloalkenyl or cycloalkenylalkyl, in each case having 3 to 8 carbon atoms in the cycloalkyl moiety or cycloalkenyl moiety, and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable substituents are: halogen, cyano and in each case straight-chain or branched alkyl or halogenoalkyl, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms, or in each case double-linked alkanediyl or alkenediyl, in each case having up to 4 carbon atoms; in addition, R^3 and R^4 independently of one another represent heterocyclylalkyl which has 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and 1 to 9 carbon atoms as well as 1 to 3 hetero atoms - in particular, nitrogen, oxygen and/or sulfur - in the heterocyclyl moiety and each of which is optionally monosubstituted or polysubstituted in the heterocyclyl moiety by identical or different substituents, whereby in each case suitable substituents are: halogen, cyano, nitro, and in each case, straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy, halogenoalkylthio or alkoxycarbonyl, in each case having 1 to 5 carbon atoms and optionally 1 to 9 identical or different halogen atoms, and in addition R^3 and R^3 independently of one another represent aralkyl, aroyl or aryl, each of which has 6 to 10 carbon atoms in the aryl moiety and optionally 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable aryl substituents are: halogen, cyano, nitro, hydroxy, in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy, halogenoalkylthio, alkylsulfinyl, alkylsulfonyl, halogenoalkylsulfinyl, halogenoalkylsulfonyl, alkanoyl or alkoxycarbonyl, in each case having 1 to 6 carbon atoms and optionally 1 to 9 identical or different halogen atoms, cycloalkyl having 3 to 6 carbon atoms or phenoxy, and whereby suitable alkyl substituents are: halogen or cyano, or
R^3 and R^4 together with the nitrogen atom to which they are bonded represent a five- to ten-membered heterocycle which can optionally contain 1 or 2 further hetero atoms, in particular, nitrogen, oxygen and/or sulfur, and which is optionally monosubstituted or polysubstituted, identically or differently, whereby suitable substituents are: halogen, and in each case straight-chain or branched alkyl or halogenoalkyl, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms as well as 1 to 2 oxo or thiono groups, whereby the compounds specially listed in the antecedent but not previously published patent application DE-P 38 09 053, in which

1. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, R^4 = cyclohexyl;
2. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, $R^4 = CH_2\text{-}C(CH_3)_3$;
3. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, R^4 = 1-phenylethyl;
4. $R^1 = C_2H_5$, $R^2 = CH_3$, $R^3 = H$, $R^4 = C(CH_3)_3$;
5. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = CH_3$, $R^4 = C(CH_3)_3$;
6. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, $R^4 = -CH(CH_3)\text{-}CH = N\text{-}OCH_3$

and

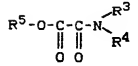
7. $R^1 = CH_3$, $R^2 = CH_3$, R^3 =H,



are excepted.

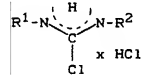
It has also been found that the novel compounds of the general formula (I) are obtained when (a) amino guanidines of the general formula (II)

in which
R^1 and R^2 have the above-mentioned meaning,
and/or tautomers of the compounds of formula (II) and/or acid adducts of compounds of formula (II) or of tautomers thereof,
are reacted with oxalic acid ester amides of the general formula (III)



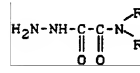
(III)

in which
R^3 and R^4 have the above-mentioned meanings and
R^5 represents alkyl,
optionally in the presence of a diluent and optionally in the presence of a reaction auxiliary, or when
(b) chloroformamidine hydrochlorides of the general formula (IV)



(IV)

in which
R^1 and R^2 have the above-mentioned meanings,
are reacted with oxalic acid amide hydrazides of the general formula (V)

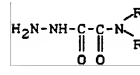


(V)

in which
R^3 and R^4 have the above-mentioned meanings,
optionally in the presence of a diluent and optionally in the presence of an acid acceptor, or when
(c) carbodiimides of the general formula (VI)

R^1-N = C= N-R^2 (VI)

in which
R^1 and R^2 have the above-mentioned meanings,
are reacted with oxalic acid amide hydrazides of the general formula (V)



(V)

in which
R^3 and R^4 have the above-mentioned meanings, optionally in the presence of a diluent and optionally in the presence of a reaction auxiliary.

Finally, it has been found that the novel 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of the general formula (I) exhibit interesting herbicidal properties.

For example, the novel 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of the general formula (I) have an excellent action against problem weeds with good to very good toleration by crop plants.

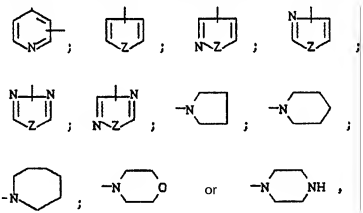
The inventive 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives are generally defined by the formula (I). Preferred compounds of formula (I) are those in which

R^1 represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n- or i-pentyl, allyl, propargyl, or represents in each case straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms, halogenoalkenyl having 3 to 6 carbon atoms or halogenoalkinyl having 3 to 6 carbon atoms and in each case 1 to 9 identical or different halogen atoms, or represents methoxymethyl, methoxyethyl,

ethoxymethyl or ethoxyethyl, or represents cyclopropyl, cyclopropylmethyl, cyclopentyl, cyclohexyl, cyclohexylmethyl, cyclohexylethyl, cyclopentylmethyl, or represents benzyl, phenylethyl or phenyl, each optionally monosubstituted to trisubstituted, identically or differently, whereby in each case suitable substituents are: fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio,

R^2 represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n- or i-pentyl or n- or i-hexyl, or represents allyl or propargyl, or represents methoxymethyl, methoxyethyl, ethoxymethyl or ethoxyethyl, or represents a straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in particular, fluorine, chlorine or bromine, or represents cyclopentyl, cyclohexyl, cyclopropyl, cyclopropylmethyl, cyclohexylmethyl or cyclohexylethyl, or represents benzyl or phenyl, each of which is optionally monosubstituted to trisubstituted by identical or different substituents, whereby in each case suitable substituents are: fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio, and R^3 and R^4 independently of one another in each case represent hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, in each case straight-chain or branched pentyl, hexyl, heptyl, octyl, nonyl, decyl or dodecyl, or represent allyl, n- or i-butenyl, n- or i-pentenyl, n- or i-hexenyl, propargyl, n- or i-butinyl, n- or i-pentinyl or n- or i-hexinyl, or represent straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, in particular, fluorine, chlorine or bromine, or represent in each case straight-chain or branched halogenoalkenyl or halogenoalkinyl, in each case having 3 to 5 carbon atoms and 1 to 3 halogen atoms, in particular, fluorine or chlorine, or represent in each case straight-chain or branched cyanoalkyl having 1 to 6 carbon atoms in the alkyl moiety, hydroxyalkyl having 1 to 6 carbon atoms and 1 to 3 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, alkylaminoalkyl or dialkylaminoalkyl, in each case having up to 4 carbon atoms in the individual alkyl or alkenyl moieties, or represent cyclopropyl, cyclopropylmethyl, cyclopropylethyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl, cyclohexylmethyl, cyclohexylethyl, cyclohexenyl, cyclohexenylmethyl or cyclohexenylethyl, each of which is optionally monosubstituted to pentasubstituted, identically or differently, whereby in each case suitable substituents are: fluorine, chlorine, bromine, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, cyano, methanediyl, ethanediyl, butanediyl or butadienediyl;

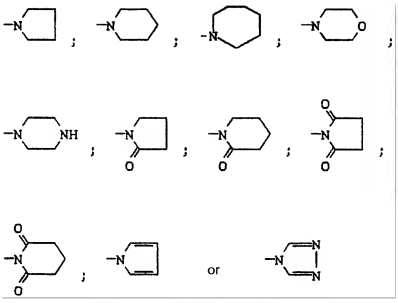
R^3 and R^4 in addition independently of one another represent heterocyclylmethyl, heterocyclylpropyl or heterocyclylethyl, each of which is optionally monosubstituted to trisubstituted in the heterocyclyl moiety by identical or different substituents, whereby in each case suitable heterocycles are:



whereby in each case Z represents oxygen or sulfur and whereby in each case suitable substituents are: fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio;

R^3 and R^4 in addition independently of one another represent benzyl, phenylethyl, phenylpropyl, phenylbutyl, phenylpentyl, phenylhexyl, phenylheptyl phenylcyanomethyl, phenylcyanoethyl, phenylcyanopropyl, benzoyl, phenyl or naphthyl, each of which is optionally straight-chain or branched in the alkyl moiety, and each of which is optionally monosubstituted to trisubstituted, identically or differently, whereby in each case suitable phenyl substituents are: fluorine, chlorine, bromine, hydroxy, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy, trifluoromethylthio, trifluoromethylsulfinyl, trifluoromethylsulfonyl, methylsulfinyl, methylsulfonyl, acetyl, propionyl, methoxycarbonyl, ethoxycarbonyl, cyclohexyl or phenoxy, or

R^3 and R^4 together with the nitrogen atom to which they are bonded represent a heterocycle of the formula



each of which is optionally monosubstituted to trisubstituted, identically or differently, whereby in each case suitable substituents are: methyl, ethyl, n- or i-propyl, chloride or trifluoromethyl, with the exception of the compounds excluded above by the disclaimer.

Particularly preferred compounds of formula (I) are those in which

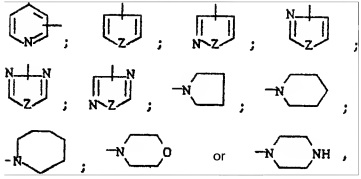
R^1 represents methyl, ethyl, propyl, isopropyl or cyclohexyl,

R^2 represents methyl, ethyl, propyl, isopropyl or cyclohexyl,

R^3 represents hydrogen or methyl,

R^4 represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n-, i-, s- or t-pentyl, n- or i-hexyl, n- or i-heptyl, n- or i-octyl, n- or i-nonyl, n- or i-decyl, n- or i-dodecyl, 1-ethylpropyl, 1,2-dimethylpropyl, 1,3-dimethylbutyl, 1-methyl-1-ethylpropyl, 1,1,3,3-tetramethylbutyl or 1,2,2-trimethylpropyl, or represents allyl, n- or i-butenyl, n- or i-pentenyl, n- or i-hexenyl, propargyl, n- or i-butinyl, n- or i-pentinyl or n- or i-hexinyl, or represents straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, in particular, fluorine or chlorine, or represents in each case straight-chain or branched halogenoalkenyl or halogenoalkinyl, in each case having 3 to 5 carbon atoms and 1 to 3 halogen atoms, in particular, fluorine or chlorine, or represents in each case straight-chain or branched cyanoalkyl having 1 to 6 carbon atoms in the alkyl moiety, hydroxyalkyl having 1 to 6 carbon atoms and 1 to 3 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, alkylaminoalkyl or dialkylaminoalkyl, in each case having up to 4 carbon atoms in the individual alkyl or alkenyl moieties, or represents cyclopropyl, cyclopropylmethyl, cyclopropylethyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl, cyclohexylmethyl, cyclohexylethyl, cyclohexenyl, cyclohexenylmethyl or cyclohexenylethyl, each of which is optionally monosubstituted to tetrasubstituted by the identical or different substituents fluorine, chlorine, bromine, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, cyano, methanediyl, ethanediyl, butanediyl or butadienediyl;

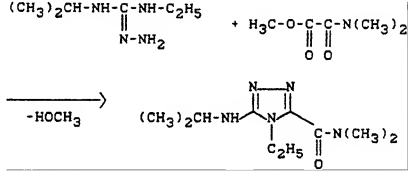
R^4 in addition represents heterocyclylmethyl, heterocyclylpropyl or heterocyclylethyl, each of which is optionally monosubstituted to trisubstituted in the heterocyclyl moiety by the identical or different substituents fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio, whereby suitable heterocycles in each case are:



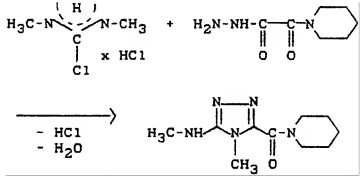
whereby in each case Z represents oxygen or sulfur,

R^4 in addition represents benzyl, phenylethyl, phenylpropyl, phenylbutyl, phenylpentyl, phenylhexyl, phenylheptyl, phenylcyanomethyl, phenylcyanoethyl, phenylcyanopropyl, benzoyl, phenyl or naphthyl, each of which is optionally straight-chain or branched in the alkyl moiety, and each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by the identical or different substituents fluorine, chlorine, bromine, hydroxy, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy, trifluoromethylthio, trifluoromethylsulfinyl, trifluoromethylsulfonyl, methylsulfinyl, methylsulfonyl, acetyl, propionyl, methoxycarbonyl, ethoxycarbonyl, cyclohexyl or phenoxy, and R^4 in addition together with R^3 can represent tetramethylene or pentamethylene, with the exception of the compounds excluded above by the disclaimer.

If, for example, 2-amino-1-ethyl-3-isopropylguanidine and oxalic acid methyl ester dimethylamide are used as starting substances, the course of the reaction in the inventive method (a) can be represented by the following equation:

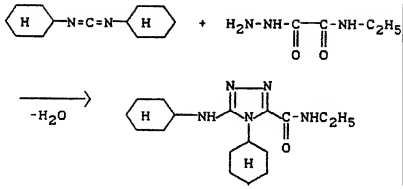


If, for example, chlorodimethyl formamidine hydrochloride and oxalic acid hydrazide piperidine are used as starting substances, the course of the reaction in the inventive method (b) can be represented by the following equation:



If, for example, dicyclohexylcarbodiimide and oxalic acid hydrazide ethylamide are used as starting substances, the course of the reaction in the inventive method (c) can be represented by the

following equation:



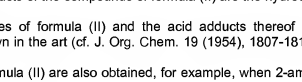
Formula (II) generally defines the aminoguanidines to be used as starting substances in the inventive method (a) for the preparation of compounds of formula (I).

In formula (II), R^1 and R^2 preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the inventive compounds of formula (I) as being preferred, or particularly preferred, for R^1 and R^2.

The preferred acid adducts of the compounds of formula (II) are the hydrochlorides, hydrobromides or hydroiodides thereof.

The starting substances of formula (II) and the acid adducts thereof are known and/or can be prepared by methods known in the art (cf. J. Org. Chem. 19 (1954), 1807-1814; Bull. Soc. Chim. France 1975, 1649-1653).

The compounds of formula (II) are also obtained, for example, when 2-amino-1,1-dimethyl-guanidine derivatives of the general formula (VII)



(VII)

in which R^1 has the above-mentioned meaning,
or tautomers of the compounds of formula (VII) and/or acid adducts (preferably hydrochlorides, hydrobromides or hydroiodides) of compounds of formula (VII) or of tautomers thereof,
are reacted with amines of the general formula (VIII)

H_2N-R^2 (VIII)

in which
R^2 has the above-mentioned meaning,
in the presence of a diluent such as, for example, isopropanol, at temperatures between 20°C and 120°C (cf. the preparation examples).

Formula (III) generally defines the oxalic acid ester amides additionally to be used as starting substances in the inventive method (a).

In formula (III), R^3 and R^4 preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the inventive compounds of the formula (I) as being preferred, or particularly preferred, for R^3 and R^4, and R^5 preferably represents C_1-C_4-alkyl, in particular, methyl or ethyl.

The starting substances of formula (III) are known and/or can be prepared by methods known in the art (cf. Unexamined Patent Application DE 28 19 878).

Formula (IV) generally defines the chloroformamidine hydrochlorides to be used as starting substances in the inventive method (b) for the preparation of compounds of formula (I).

In formula (IV), R^1 and R^2 preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the inventive compounds of the formula (I) as

being preferred, or particularly preferred, for R^1 and R^2.

The starting substances of formula (IV) are known and/or can be prepared by methods known in the art (cf. Unexamined Patent Application DE 37 09 574, Chem. Ber. 97 (1964), 1232-1245) - cf. also the preparation examples.

Formula (V) generally defines the oxalic acid amide hydrazides to be used as starting substances in the inventive methods (b) and (c).

In formula (V), R^3 and R^4 preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the inventive compounds of the formula (I) as being preferred, or particularly preferred, for R^3 and R^4.

The starting substances of formula (V) are known and/or can be prepared by methods known in the art (cf. EP-A 126 326).

Formula (VI) generally defines the carbodiimides to be used as starting substances in the inventive method (c) for the preparation of compounds of formula (I).

In formula (VI), R^1 and R^2 preferably, or in particular, have those meanings which have already been mentioned above in connection with the description of the inventive compounds of the formula (I) as being preferred, or particularly preferred, for R^1 and R^2.

The starting substances of formula (VI) are known organic synthesis chemicals.

Suitable diluents for carrying out the inventive method (a) are inert organic solvents. These include, in particular, aliphatic, alicyclic or aromatic, optionally halogenated hydrocarbons such as, for example, benzine, benzene, toluene, xylene, chlorobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, chloroform, carbon tetrachloride, ethers such as diethyl ether, dioxane, tetrahydrofuran, ethylene glycol dimethyl ether or ethylene glycol diethyl ether, nitriles such as acetonitrile or propionitrile, amides such as dimethylformamide, dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide, or alcohols such as methanol, ethanol or propanol.

The inventive method (a) is optionally carried out in the presence of a suitable reaction auxiliary. Suitable reaction auxiliaries are all inorganic and organic bases that can typically be used. Preferably used are the alkali metal hydrides, alkali metal hydroxides, alkali metal amides, alkali metal alcoholates, alkali metal carbonates or alkali metal hydrogen carbonates such as, for example, sodium hydride, sodium amide, sodium hydroxide, sodium methylate, sodium ethylate, potassium t-butylate, sodium carbonate or sodium hydrogen carbonate, or also tertiary amines such as, for example, triethylamine, N,N-dimethylaniline, pyridine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

When carrying out the inventive method (a), the reaction temperatures can be varied within a large range. In general, temperatures between 30°C and 150°C are used, preferably temperatures between 50°C and 80°C.

To carry out the inventive method (a), 0.8 to 1.5 moles, preferably 0.8 to 1.2 moles, of oxalic ester amide of formula (III) and optionally 1 to 5 moles, preferably 1 to 2.5 moles, of reaction auxiliary are generally employed per mole of aminoguanidine of formula (II) or of a corresponding acid addition salt.

The reaction is carried out and the reaction products are worked up and isolated using generally prevailing methods (cf. the preparation examples as well).

The inventive method (b) is optionally carried out in the presence of a diluent. Preferably employed as diluents are polar organic solvents and/or water. Preferred organic solvents are alcohols such as methanol, ethanol, propanol, isopropanol and butanol, ethers such as ethylene glycol dimethyl ether, diethylene glycol dimethyl ether, tetrahydrofuran and dioxane, ether alcohols such as ethylene glycol monomethyl ether and ethylene glycol monoethyl ether, amides such as formamide and dimethylformamide, nitriles such as acetonitrile, propionitrile or benzonitrile, and pyridine.

Acid acceptors that can be employed in the inventive method (b) are all acid-binding agents that can typically be used for reactions of this type. Preferably used are alkali metal hydroxides such as, for example, sodium hydroxide and potassium hydroxide, alkaline earth metal hydroxides such as, for example, calcium hydroxide, alkali carbonates and alkali alcoholates such as sodium carbonate and potassium carbonate, sodium methylate and potassium methylate, and in addition aliphatic, aromatic or heterocyclic amines, for example, triethylamine, trimethylamine, dimethylaniline, dimethylbenzylamine, pyridine, 1,5-diazabicyclo-[4,3,0]-non-5-ene (DBU), 1,8-diazabicyclo-[5,4,0]-undec-7-ene (DBO) and 1,4-diazabicyclo-[2,2,2]-octane (DABCO).

When carrying out the inventive method (b), the reaction temperatures can be varied within a large range. In general, the method is carried out at temperatures between 0°C and 150°C, preferably at temperatures between 10°C and 120°C.

In general, the inventive method (b) is carried out under atmospheric pressure. However, it is also suitable to carry out the method under increased or reduced pressure.

To carry out the inventive method (b), between 0.5 and 1.5 moles, preferably between 0.8 and 1.2 moles, of oxalic acid amide hydrazide of formula (V) and between 1 and 5 mole equivalents, preferably between 2 and 3 mole equivalents, of an acid acceptor are generally employed per 1 mole of chloroformamidine hydrochloride of formula (IV).

In general, the reactants of formula (II) and (III) are mixed with the diluent at room temperature, and after an acid acceptor is added, they are then stirred until the reaction is complete, optionally at an increased temperature.

The reaction products can be worked up using conventional methods (cf. the preparation examples).

The inventive method (c) is optionally carried out in the presence of a diluent. The same diluents can be used for this purpose as have been mentioned above in the inventive methods (a) and (b).

Optionally, method (c) is carried out in the presence of a reaction auxiliary. The same reaction auxiliaries can be employed for this purpose as have been mentioned above in the inventive method (a).

When carrying out the inventive method (c), the reaction temperatures can be varied within a large range. In general, this is carried out at temperatures between 0°C and 150°C, preferably at temperatures between 20°C and 120°C.

In general, the inventive method (c) is carried out under atmospheric pressure. However, it is also suitable to carry out the method under increased or reduced pressure.

To carry out the inventive method (c), the particular required starting substances are generally employed in approximately equimolar amounts. However, it is also possible to use one of the two employed components in a large excess. In general, the reactions are carried out in a suitable diluent, and the reaction mixture is stirred for several hours at the particular required temperature. Each of the reaction products in the inventive method (c) is worked up using conventional methods (cf. the preparation examples).

The inventive active substances can be used as defoliants, desiccants, agents for destroying broad-leaved plants and, in particular, as weed-killers. The term "weeds", in the broadest sense, is understood to mean all plants that grow in locations where they are not wanted. Whether the inventive substances act as total or selective herbicides essentially depends on the amount used.

The inventive active substances can be used, for example, in connection with the following plants:

Dicotyledon weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Matricaria, Anthemis, Galinsoga, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca, Xanthium, Convolvulus, Ipomoea, Polygonum, Sesbania, Ambrosia, Cirsium, Carduus, Sonchus, Solanum, Rorippa, Rotala, Lindernia, Lamium, Veronica, Abutilon, Emex, Datura, Viola, Galeopsis, Papaver and Centaurea.

Dicotyledon cultures of the genera: Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica, Lactuca, Cucumis and Cucurbita.

Monocotyledon weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Brachiaria, Lolium, Bromus, Avena, Cyperus, Sorghum, Agropyron, Cynodon, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Sphenoclea, Dactyloctenium, Agrostis, Alopecurus and Apera.

Monocotyledon cultures of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale, Sorghum, Panicum, Saccharum, Ananas, Asparagus and Allium.

However, the use of the inventive active substances is in no way restricted to these genera, but rather extends in the same manner to other plants as well.

Depending on the concentration, the compounds are suitable for the total weed control, for example, on industrial facilities and railroad tracks, and on paths and spaces with or without tree growth. In the same way, the compounds can be employed for weed control in perennial cultures, for example, forests, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil palm plantations, cocoa plantations, berry plantings and hop fields, and for selective weed control in annual cultures.

The inventive compounds of the formula (I) are particularly suitable for selectively controlling dicotyledon weeds in monycotyledon crops such as, for example, in corn, in both the pre-emergence and the post-emergence method.

To a certain extent, the compounds of formula (I) also show a fungicidal action such as, for example, against apple scab (Venturia inaequalis).

The active substances can be converted into the usual formulations such as solutions, emulsions, wettable powders, suspensions, powders, dusting agents, pastes, soluble powders, granulates, suspension-emulsion concentrates, natural and synthetic materials impregnated with active substance, and superfine encapsulations in polymeric substances.

These formulations are produced in a known manner, for example, by mixing the active substances with extenders, i.e., liquid solvents and/or solid carriers, optionally including the use of surface-active agents, i.e., emulsifying agents and/or dispersing agents and/or foam-forming agents.

In the case of the use of water as an extender, organic solvents, for example, can also be used as auxiliary solvents. Basically, suitable liquid solvents are: aromatics such as xylene, toluene, or alkyl naphthalenes, chlorinated aromatics and chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example, petroleum fractions, mineral and vegetable oils, alcohols such as butanol or glycol and ethers and esters thereof, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide and dimethyl sulfoxide, as well as water.

Suitable as solid carriers are:

For example, ammonium salts and powdered natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and powdered synthetic minerals such as highly disperse silica, aluminum oxides and silicates, suitable as solid carriers for granulates are: for example, crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granulates of inorganic and organic powders, as well as granulates of organic material such as sawdust, coconut shells, corn cobs and tobacco stalks; suitable as emulsifying and/or foam-forming agents are: for example, nonionic and anionic emulsifiers such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example, alkylaryl polyglycol ethers, alkyl sulfonates, alkyl sulfates, aryl sulfonates, as well as albumin hydrolysis products; suitable as dispersing agents are: for example, lignin-sulfite waste liquors and methylcellulose.

Adhesives such as carboxymethylcellulose, natural and synthetic powder, granulate or latex polymers such as gum arabic, polyvinyl alcohol and polyvinyl acetate can be used in the formulations, as well as natural phospholipids such as cephalins and lecithins, and synthetic phospholipids. Further additives can be mineral and vegetable oils.

Dyes such as inorganic pigments, for example, iron oxide, titanium oxide and ferrocyanide blue, and organic dyes such as alizarin dyes, azo dyes and metal phthalocyanine dyes, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc can be used.

The formulations generally contain between 0.1 and 95 weight-percent active substance, preferably between 0.5 and 90%.

The inventive active substances, as such or in the form of formulations thereof, can also be used in mixtures with known herbicides for weed control, whereby finished formulations or tank mixtures are suitable.

Known herbicides for the mixtures are, for example, 1-amino-6-ethylthio-3-(2,2-dimethylpropyl)-1,3,5-triazine-2,4(1H,3H)-dione (AMETHYDIONE) or N-(2-benzothiazolyl)-N,N'-dimethyl-urea (METABENZTHIAZURON) for weed control in grains; 4-amino-3-methyl-6-phenyl-1,2,4-triazin-5(4H)-one (METAMITRON) for weed control in sugar beets, and 4-amino-6-(1,1-dimethylethyl)-3-methylthio-1,2,4-triazin-5(4H)-one (METRIBUZIN) for weed control in soybeans; in addition, also 2,4-dichlorophenoxyacetic acid (2,4-D); 4-(2,4-dichlorophenoxy)-butyric acid (2,4-DB); chloroacetic acid-N-(methoxymethyl)-2,6-diethylanilide (ALACHLOR); 2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine (ATRAZINE); 3-isopropyl-2,1,3-benzothiadiazin-4-one-2,2-dioxide (BENTAZONE); 3,5-dibromo-4-hydroxy-benzonitrile (BROMOXYNIL); 2-chloro-4-ethylamino-6-(3-cyanopropylamino)-1,3,5-triazine (CYANAZINE); 4-amino-6-t-butyl-3-ethylthio-1,2,4-triazin-5(4H)-one (ETHIOZIN); 3,5-diiodo-4-hydroxybenzonitrile (IOXYNIL); N-methyl-2-(1,3-benzothiazol-2-yloxy)-acetanilide (MEFENACET); 2-chloro-N-(2,6-dimethylphenyl)-N-[(1H)-pyrazol-1-yl-methyl]-acetamide (METAZACHLOR); 2-ethyl-6-methyl-N-(1-methyl-2-methoxyethyl)-chloroacetanilide (METOLACHLOR); N-(1-ethylpropyl)-3,4-dimethyl-2,6-dinitroaniline (PENDIMETHALIN); 0-(6-chloro-3-phenylpyridazin-4-yl)-S-octyl-thiocarbonate (PYRIDATE) and 3-[[[[(4-methoxy-6-methyl-1,3,5-(triazin-2-yl)-amino]-carbonyl]-amino]-sulfonyl]-thiophene-2-carboxylic acid methyl ester (THIAMETURON). Surprisingly, some mixtures also show synergistic action.

Also suitable are mixture with other known active substances such as fungicides, insecticides, acaricides, nematicides, bird repellents, plant nutrients and agents that improve soil structure.

The active substances can be used as such, in the form of formulations thereof, or in the usage forms prepared therefrom by further dilution, such as ready-to-use solutions, suspensions, emulsions, powders, pastes and granulates. Usage takes place in the usual manner, for example, by watering, spraying, atomizing or spreading.

The inventive active substances can be applied either before or after emergence of

the plants.

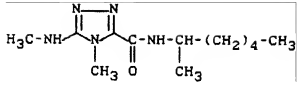
They can also be incorporated into the soil before sowing.

The amount of active substance used can vary within a large range. It essentially depends on the nature of the desired effect. In general, the amounts used are between 0.01 and 10 kg of active substance per hectare of land area, preferably between 0.05 and 5 kg per ha.

The preparation and use of the inventive active substances can be seen from the following examples.

Preparation Examples:

Example 1

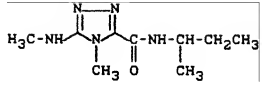


(Method (a))

A mixture of 24.8 g (0.18 mol) 2-amino-1,3-dimethyl-guanidine hydrochloride, 32.9 g (0.15 mol) oxalic acid ethyl ester sec-heptylamide, 16.2 g (0.3 mol) sodium methylate and 200 ml methanol is stirred for 3 hours at reflux temperature. The mixture is subsequently cooled to room temperature and filtered. The filtrate is concentrated in a water jet vacuum, the residue is taken up in 200 ml dichloromethane, washed three times, each time with 100 ml water, dried over sodium sulfate and filtered. The solvent is distilled from the filtrate in a water jet vacuum.

This yields 24.8 g (65% of theory) 5-methylamino-4-methyl-4H-1,2,4-triazol-3-yl-carboxylic acid-sec-heptylamide with a melting point of 101°C-103°C.

Example 2

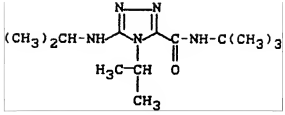


(Method (b))

A mixture of 7.15 g (0.05 mol) chloro-N,N'-dimethylformamidine hydrochloride, 7.9 g (0.05 mol) oxalic acid hydrazide-sec-butylamide, 5.4 g (0.1 mol) sodium methylate and 200 ml butanol is stirred at reflux temperature for 60 minutes. The mixture is subsequently cooled to room temperature and filtered. The filtrate is concentrated in a water jet vacuum, the residue is taken up in 150 ml of dichloromethane, washed three times, each time with 100 ml water, dried over sodium sulfate and filtered. The solvent is distilled from the filtrate in a water jet vacuum.

This yields 4.4 g (40% of theory) 5-methylamino-4-methyl-4H-1,2,4-triazol-3-yl-carboxylic acid-sec-butylamide with a melting point of 129°C-131°C.

Example 3

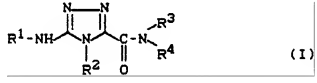


(Method (c))

A mixture of 12.6 g (0.1 mol) N,N'-diisopropylcarbodiimide, 15.9 g (0.1 mol) oxalic acid-hydrazide-tert-butylamide, 0.8 g sodium methylate and 200 ml butanol is stirred for 3 hours at reflux temperature, then cooled to room temperature and filtered. The filtrate is concentrated in a water jet vacuum, the residue is taken up in 200 ml of dichloromethane, washed three times, each time with 100 ml water, dried over sodium sulfate and filtered. The solvent is distilled from the filtrate in a water jet vacuum. The raw product obtained as the residue is purified by means of column chromatography (silica gel; cyclohexane/ethanol 1 :1) and recrystallized from cyclohexane.

This yields 8.7 g (33% of theory) 5-isopropylamino-4-isopropyl-4H-1,2,4-triazol-3-yl-carboxylic acid-tert-butylamide with a melting point of 135-137°C.

Analogous to Examples 1 to 3 and in accordance with the general description of the inventive preparation methods, the compounds of formula (I) listed in Table 1 below can also be prepared, for example.



(I)

Table 1: Examples of the compounds of formula (I)

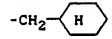
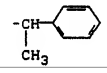
Example No.	R^1	R^2	R^3	R^4	Melting Point (°C)
4	CH_3	CH_3	H	$-C(CH_3)_3$	166-167
5	CH_3	CH_3	H	$-CH(C_2H_5)_2$	141-143
6	CH_3	CH_3	H	$-\underset{\displaystyle CH_3}{\overset{|}{CH}}-CH(CH_3)_2$	130-132
7	CH_3	CH_3	H	$-CH_2CH=CH_2$	125-127
8	CH_3	CH_3	H	$-CH(CH_3)_2$	122-124
9	CH_3	CH_3	H		132-133
10	CH_3	CH_3	H		146-148
11	CH_3	CH_3	H		118-120

Table 1: - Continued

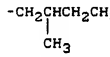
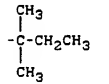
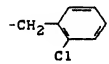
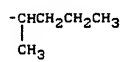
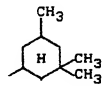
Example No.	R^1	R^2	R^3	R^4	Melting Point (°C)
12	CH_3	CH_3	H		159-160
13	CH_3	CH_3	H	$-CH_2CH_2Cl$	221-223
14	CH_3	CH_3	H		160-161
15	CH_3	CH_3	H		79-81
16	CH_3	CH_3	H	$-CH_2CH(CH_3)_2$	116-118
17	CH_3	CH_3	H		150-152
18	CH_3	CH_3	H		219-221
19	CH_3	CH_3	H		150-152
20	CH_3	CH_3	H		118-120
21	CH_3	CH_3	H		152

Table 1: - Continued

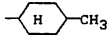
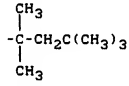
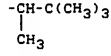
Example No.	R^1	R^2	R^3	R^4	Melting Point (°C)
22	CH_3	CH_3	H		137-139
23	CH_3	CH_3	H		132-134
24	CH_3	CH_3	H		115-117
25	CH_3	CH_3	H		97-99
26	CH_3	CH_3	H		124-126
27	CH_3	CH_3	H		90-92
28	CH_3	CH_3	H		191-193

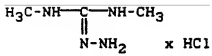
Table 1: - Continued

Example No.	R^1	R^2	R^3	R^4	Melting Point (°C)
29	CH_3	CH_3	H		109-111
30	CH_3	CH_3	H	$-CH(CH_3)-CH_2-CH(CH_3)_2$	114-116
31	CH_3	CH_3	H	$-CH_2CH_2CH_2CH_3$	118-120
32	CH_3	CH_3	H	$-C(CH_3)_3$	245-247
33			H	$-C(CH_3)_3$	250
34	CH_3	CH_3	—$(CH_2)_5$—		*)

*) ^{1}H-NMR (DMSO-D^6, δ, ppm): 1,50-1,65; 2,80; 6,25.

Starting Substances of Formula (II):

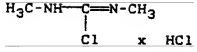
Example (II-1)



152.5 g (1.0 mol) 2-amino-1,1,3-trimethylguanidine hydrochloride is heated to reflux temperature in 2.0 liters isopropanol. In conjunction with that, 155 g (5 mol) methylamine is introduced within 2 hours. The mixture is cooled to 15°C, and the product obtained in crystalline form is then isolated by means of suction.
This yields 98.2 [g] (71% of theory) 2-amino-1,3-dimethyl-guanidine hydrochloride with a melting point of 250°C.
^{1}H-NMR (DMSO-D^6, δ, ppm): 2.7-2.8.

Starting Substances of Formula (IV)

Example (IV-1)



110 g (1.1 mol) phosgene is introduced at 80°C within 1.5 hours into a mixture of 88 g (1 mol) N,N'-dimethylurea and 500 ml chlorobenzene, and, after this introduction has been completed, stirring is continued for an additional 45 minutes at 80°C until the carbon dioxide development has ended. The reaction mixture is cooled under nitrogen, and the solids are suctioned off under nitrogen at 20°C. To purify the solid product that is obtained following concentration of the filtrate, it is dissolved in chloroform and precipitated with tetrahydrofuran.
This yields 34 g (24% of theory) N,N'-dimethylchloroformamidine hydrochloride with a melting point of 156°C-158°C.

Examples of Use

Example A

Pre-emergence Test

Solvent: 5 parts by weight acetone
Emulsifier:1 part by weight alkylaryl polyglycol ether
To produce a useful active substance preparation, 1 part by weight active substance is mixed with the indicated amount of solvent, the indicated amount of emulsifier is added, and the concentrate is diluted with water to the desired concentration.
Seeds of the test plants are sown in normal soil and are watered with the preparation of active substance 24 hours later. It is expedient in this regard to keep the amount of water per unit area constant. The concentration of active substance in the preparation is not important, only the amount of active substance used per unit area is critical. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control. The meanings are as follows:
0% = No action (like the untreated control)
100% = Total destruction
In this test, the compounds of preparation examples 2, 4, 5, 6, 8, 9, 11, 12, 15, 16, 17, 20, 22, 23 and 24, for example, show excellent effectiveness in weed control, in particular, dicotyledon weeds, combined with a good selectivity in crop plants.

Example B

Post-emergence test

Solvent: 5 parts by weight acetone

Emulsifier: 1 part by weight alkylaryl polyglycol ether

To produce a useful active substance preparation, 1 part by weight active substance is mixed with the indicated amount of solvent, the indicated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Test plants with a height of 5-15 cm are sprayed with the active substance preparation in such a way that the particular desired amounts of active substance per unit area are applied. The concentration of the spray liquor is chosen so that the particular desired amounts of active substance are applied in 2,000 liters of water/ha. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control.

The meanings are as follows:

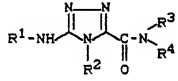
0% = No action (like the untreated control)

100% = Total destruction

In this test, the compounds of preparation examples 1, 2, 4, 5, 6, 7, 8, 9, 10, 11, 12, 14, 15, 16, 17, 18, 19, 20, 22, 23, 24 and 34, for example, show excellent herbicidal action in weed control, in particular, dicotyledon weeds, combined with good selectivity in crop plants.

Claims:

1. 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of the general formula (I)



(I)

in which

R^1 represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl or halogenoalkinyl having in each case 2 to 8 carbon atoms and 1 to 15, or 13 respectively, identical or different halogen atoms, alkoxyalkyl having 1 to 6 carbon atoms in the individual alkyl moieties, or represents cycloalkyl having 3 to 7 carbon atoms, or represents cycloalkylalkyl having 3 to 7 carbon atoms in the cycloalkyl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, or represents aralkyl which has 6 to 10 carbon atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 3 hetero atoms, in particular, nitrogen, oxygen and/or sulfur, whereby in each case suitable substituents are: halogen, cyano, nitro and in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy or halogenoalkylthio, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms,

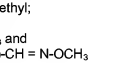
R^2 represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl having 2 to 8 carbon atoms and 1 to 15 identical or different halogen atoms, halogenoalkinyl having 2 to 8 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxyalkyl having 1 to 6 carbon atoms in each of the individual alkyl moieties, or represents cycloalkylalkyl or cycloalkyl, in each case having 3 to 7 carbon atoms in the cycloalkyl moiety and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, or represents aralkyl or aryl, each of which has 6 to 10 carbon atoms in the aryl moiety and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable aryl substituents are: halogen, cyano, nitro and in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy or halogenoalkylthio, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms, and

R^3 and R^4 independently of one another in each case represent hydrogen, or in each case represent straight-chain or branched alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl

having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl or halogenoalkinyl, in each case having 2 to 8 carbon atoms and 1 to 15, or 13 respectively, identical or different halogen atoms, cyanoalkyl having 1 to 8 carbon atoms, hydroxyalkyl having 1 to 8 carbon atoms and 1 to 6 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, in each case having up to 6 carbon atoms in the individual alkyl or alkenyl moieties, alkylaminoalkyl or dialkylaminoalkyl, in each case having 1 to 6 carbon atoms in the individual alkyl moieties, or cycloalkyl, cycloalkylalkyl, cycloalkenyl or cycloalkenylalkyl, in each case having 3 to 8 carbon atoms in the cycloalkyl moiety or cycloalkenyl moiety, and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable substituents are: halogen, cyano and in each case straight-chain or branched alkyl or halogenoalkyl, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms, or in each case double-linked alkanediyl or alkenediyl, in each case having up to 4 carbon atoms; in addition, R^3 and R^4 independently of one another represent heterocyclylalkyl which has 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and 1 to 9 carbon atoms as well as 1 to 3 hetero atoms - in particular, nitrogen, oxygen and/or sulfur - in the heterocyclyl moiety and each of which is optionally monosubstituted or polysubstituted in the heterocyclyl moiety by identical or different substituents, whereby in each case suitable substituents are: halogen, cyano, nitro, and in each case, straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy, halogenoalkylthio or alkoxycarbonyl, in each case having 1 to 5 carbon atoms and optionally 1 to 9 identical or different halogen atoms, and in addition R^3 and R^4 independently of one another represent aralkyl, aroyl or aryl, each of which has 6 to 10 carbon atoms in the aryl moiety and optionally 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable aryl substituents are: halogen, cyano, nitro, hydroxy, in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy, halogenoalkylthio, alkylsulfinyl, alkylsulfonyl, halogenoalkylsulfinyl, halogenoalkylsulfonyl, alkanoyl or alkoxycarbonyl, in each case having 1 to 6 carbon atoms and optionally 1 to 9 identical or different halogen atoms, cycloalkyl having 3 to 6 carbon atoms or phenoxy, and whereby suitable alkyl substituents are: halogen or cyano, or

R^3 and R^4 together with the nitrogen atom to which they are bonded represent a five- to ten-membered heterocycle which can optionally contain 1 or 2 further hetero atoms, in particular, nitrogen, oxygen and/or sulfur, and which is optionally monosubstituted or polysubstituted, identically or differently, whereby suitable substituents are: halogen, and in each case straight-chain or branched alkyl or halogenoalkyl, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms as well as 1 to 2 oxo or thiono groups, whereby the compounds in which

1. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, R^4 = cyclohexyl;
2. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, $R^4 = CH_2\text{-}C(CH_3)_3$;
3. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, R^4 = 1-phenylethyl;
4. $R^1 = C_2H_5$, $R^2 = CH_3$, $R^3 = H$, $R^4 = C(CH_3)_3$;
5. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = CH_3$, $R^4 = C(CH_3)_3$ and
6. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, $R^4 = \text{-}(CH(CH_3)\text{-}CH = N\text{-}OCH_3$
7. $R^1 = CH_3$, $R^2 = CH_3$, R^3 =H,

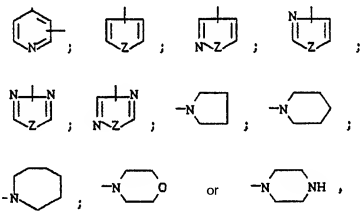


are excepted.

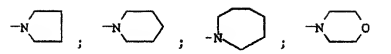
2. 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of formula (I) according to claim 1, in which

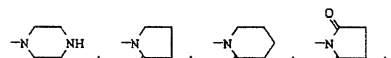
R^1 represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n- or i-pentyl, allyl, propargyl, or represents in each case straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms, halogenoalkenyl having 3 to 6 carbon atoms or halogenoalkinyl having 3 to 6 carbon atoms and in each case 1 to 9 identical or different halogen atoms, or represents methoxymethyl, methoxyethyl, ethoxymethyl or ethoxyethyl, or represents cyclopropyl, cyclohexylmethyl [sic], cyclopentyl, cyclohexyl, cyclohexylmethyl, cyclohexylethyl, cyclopentylmethyl, or represents benzyl, phenylethyl or phenyl, each optionally monosubstituted to trisubstituted, identically or differently, whereby in each case suitable substituents are: fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or

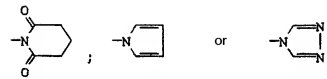
trifluoromethylthio,
R^2 represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n- or i-pentyl or n- or i-hexyl, or represents allyl or propargyl, or represents methoxymethyl, methoxyethyl, ethoxymethyl or ethoxyethyl, or represents a straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or represents cyclopentyl, cyclohexyl, cyclopropyl, cyclopropylmethyl, cyclohexylmethyl or cyclohexylethyl, or represents benzyl or phenyl, each of which is optionally monosubstituted to trisubstituted by identical or different substituents, whereby in each case suitable substituents are: fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio, and
R^3 and R^4 independently of one another in each case represent hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, in each case straight-chain or branched pentyl, hexyl, heptyl, octyl, nonyl, decyl or dodecyl, or represent allyl, n- or i-butenyl, n- or i-pentenyl, n- or i-hexenyl, propargyl, n- or i-butinyl, n- or i-pentinyl or n- or i-hexinyl, or represent straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, or represent in each case straight-chain or branched halogenoalkenyl or halogenoalkinyl, in each case having 3 to 5 carbon atoms and 1 to 3 halogen atoms, or represent in each case straight-chain or branched cyanoalkyl having 1 to 6 carbon atoms in the alkyl moiety, hydroxyalkyl having 1 to 6 carbon atoms and 1 to 3 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, alkylaminoalkyl or dialkylaminoalkyl, in each case having up to 4 carbon atoms in the individual alkyl or alkenyl moieties, or represent cyclopropyl, cyclopropylmethyl, cyclopropylethyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl, cyclohexylmethyl, cyclohexylethyl, cyclohexenyl, cyclohexenylmethyl or cyclohexenylethyl, each of which is optionally monosubstituted to pentasubstituted, identically or differently, whereby in each case suitable substituents are: fluorine, chlorine, bromine, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, cyano, methanediyl, ethanediyl, butanediyl or butadienediyl;
R^3 and R^4 in addition independently of one another represent heterocyclylmethyl, heterocyclylpropyl or heterocyclylethyl, each of which is optionally monosubstituted to trisubstituted in the heterocyclyl moiety by identical or different substituents, whereby in each case suitable heterocycles are:



whereby in each case Z represents oxygen or sulfur and whereby in each case suitable substituents are: fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio;
R^3 and R^4 in addition independently of one another represent benzyl, phenylethyl, phenylpropyl, phenylbutyl, phenylpentyl, phenylhexyl, phenylheptyl, phenylcyanomethyl, phenylcyanoethyl phenylcyanopropyl, benzoyl, phenyl or naphthyl, each of which is optionally straight-chain or branched in the alkyl moiety, and each of which is optionally monosubstituted to trisubstituted, identically or differently, whereby in each case suitable phenyl substituents are: fluorine, chlorine, bromine, hydroxy, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy, trifluoromethylthio, trifluoromethylsulfinyl, trifluoromethylsulfonyl, methylsulfinyl, methylsulfonyl, acetyl, propionyl, methoxycarbonyl, ethoxycarbonyl, cyclohexyl or phenoxy, or
R^3 and R^4 together with the nitrogen atom to which they are bonded represent a heterocycle of the formula





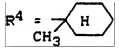


each of which is optionally monosubstituted to trisubstituted, identically or differently, whereby in each case suitable substituents are: methyl, ethyl, n- or i-propyl, chloride or trifluoromethyl, whereby the compounds in which

1. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, R^4 = cyclohexyl;
2. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, $R^4 = CH_2\text{-}C(CH_3)_3$;
3. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, R^4 = 1-phenylethyl;
4. $R^1 = C_2H_5$, $R^2 = CH_3$, $R^3 = H$, $R^4 = C(CH_3)_3$;
5. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = CH_3$, $R^4 = C(CH_3)_3$;
6. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, $R^4 = \text{-}9CH(CH_3)\text{-}CH = N\text{-}OCH_3$

and

7. $R^1 = CH_3$, $R^2 = CH_3$, R^3 =H,



are excepted.

3. 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of formula (I) according to claim 1, in which

R^1 represents methyl, ethyl, propyl, isopropyl or cyclohexyl,

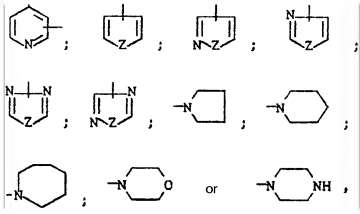
R^2 represents methyl, ethyl, propyl, isopropyl or cyclohexyl,

R^3 represents hydrogen or methyl,

R^4 represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, n-, i-, s- or t-pentyl, n- or i-hexyl, n- or i-heptyl, n- or i-octyl, n- or i-nonyl, n- or i-decyl, n- or i-dodecyl, 1-ethylpropyl, 1,2-dimethylpropyl, 1,3-dimethylbutyl, 1-methyl-1-ethylpropyl, 1,1,3,3-trimethylbutyl or 1,2,2-trimethylpropyl, or represents allyl, n- or i-butenyl, n- or i-pentenyl, n- or i-hexenyl, propargyl, n- or i-butinyl, n- or i-pentinyl or n- or i-hexinyl, or represents straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 9 identical or different halogen atoms, in particular, fluorine or chlorine, or represents in each case straight-chain or branched halogenoalkenyl or halogenoalkinyl, in each case having 3 to 5 carbon atoms and 1 to 3 halogen atoms, in particular, fluorine or chlorine, or represents in each case straight-chain or branched cyanoalkyl having 1 to 6 carbon atoms in the alkyl moiety, hydroxyalkyl having 1 to 6 carbon atoms and 1 to 3 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, alkylaminoalkyl or dialkylaminoalkyl, in each case having up to 4 carbon atoms in the individual alkyl or alkenyl moieties, or represents cyclopropyl, cyclopropylmethyl, cyclopropylethyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl, cyclooctyl, cyclohexylmethyl, cyclohexylethyl, cyclohexenyl, cyclohexenylmethyl or cyclohexenylethyl, each of which is optionally monosubstituted to tetrasubstituted by the identical or different substituents fluorine, chlorine, bromine, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, cyano, methanediyl, ethanediyl, butanediyl or butadienediyl;

R^4 in addition represents heterocyclylmethyl, heterocyclylpropyl or heterocyclylethyl, each of which is optionally monosubstituted to trisubstituted in the heterocyclyl moiety by the identical or different substituents fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl,

methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy or trifluoromethylthio, whereby suitable heterocycles in each case are:



whereby in each case Z represents oxygen or sulfur,

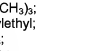
R^4 in addition represents benzyl, phenylethyl, phenylpropyl, phenylbutyl, phenylpentyl, phenylhexyl, phenylheptyl, phenylcyanomethyl, phenylcyanoethyl, phenylcyanopropyl, benzoyl, phenyl or naphthyl, each of which is optionally straight-chain or branched in the alkyl moiety, and each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by the identical or different substituents fluorine, chlorine, bromine, hydroxy, cyano, nitro, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, methylthio, trifluoromethyl, trifluoromethoxy, trifluoromethylthio, trifluoromethylsulfinyl, trifluoromethylsulfonyl, methylsulfinyl, methylsulfonyl, acetyl, propionyl, methoxycarbonyl, ethoxycarbonyl, cyclohexyl or phenoxy, and R^4 in addition together with R^3 can represent tetramethylene or pentamethylene,

whereby the compounds in which

1. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, R^4 = cyclohexyl;
2. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, $R^4 = CH_2\text{-}C(CH_3)_3$;
3. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, R^4 = 1-phenylethyl;
4. $R^1 = C_2H_5$, $R^2 = CH_3$, $R^3 = H$, $R^4 = C(CH_3)_3$;
5. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = CH_3$, $R^4 = C(CH_3)_3$;
6. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, $R^4 = \text{-}CH(CH_3)\text{-}CH = N\text{-}OCH_3$

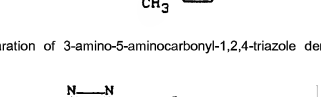
and

7. $R^1 = CH_3$, $R^2 = CH_3$, R^3 =H,



are excepted.

4. Method for the preparation of 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of the general formula (I)



(I)

in which

R^1 represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl or halogenoalkinyl having in each case 2 to 8 carbon atoms and 1 to 15, or 13 respectively, identical or different halogen atoms, alkoxyalkyl having 1 to 6 carbon atoms in the individual alkyl moieties, or represents cycloalkyl having 3 to 7 carbon atoms, or represents cycloalkylalkyl having 3 to 7 carbon atoms in the cycloalkyl moiety and 1 to 6 carbon atoms in

the straight-chain or branched alkyl moiety, or represents aralkyl which has 6 to 10 carbon atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 3 hetero atoms, in particular, nitrogen, oxygen and/or sulfur, whereby in each case suitable substituents are: halogen, cyano, nitro and in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy or halogenoalkylthio, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms,

R^2 represents in each case straight-chain or branched alkyl having 1 to 8 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl having 2 to 8 carbon atoms and 1 to 15 identical or different halogen atoms, halogenoalkinyl having 2 to 8 carbon atoms and 1 to 13 identical or different halogen atoms, alkoxyalkyl having 1 to 6 carbon atoms in each of the individual alkyl moieties, or represents cycloalkylalkyl or cycloalkyl, in each case having 3 to 7 carbon atoms in the cycloalkyl moiety and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, or represents aralkyl or aryl, each of which has 6 to 10 carbon atoms in the aryl moiety and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable aryl substituents are: halogen, cyano, nitro and in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy or halogenoalkylthio, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms, and

R^3 and R^4 independently of one another in each case represent hydrogen, or in each case represent straight-chain or branched alkyl having 1 to 18 carbon atoms, alkenyl having 2 to 8 carbon atoms, alkinyl having 2 to 8 carbon atoms, halogenoalkyl having 1 to 8 carbon atoms and 1 to 17 identical or different halogen atoms, halogenoalkenyl or halogenoalkinyl, in each case having 2 to 8 carbon atoms and 1 to 15, or 13 respectively, identical or different halogen atoms, cyanoalkyl having 1 to 8 carbon atoms, hydroxyalkyl having 1 to 8 carbon atoms and 1 to 6 hydroxy groups, alkoxyalkyl, alkoximinoalkyl, alkoxycarbonylalkyl or alkoxycarbonylalkenyl, in each case having up to 6 carbon atoms in the individual alkyl or alkenyl moieties, alkylaminoalkyl or dialkylaminoalkyl, in each case having 1 to 6 carbon atoms in the individual alkyl moieties, or cycloalkyl, cycloalkylalkyl, cycloalkenyl or cycloalkenylalkyl, in each case having 3 to 8 carbon atoms in the cycloalkyl moiety or cycloalkenyl moiety, and optionally 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety, each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable substituents are: halogen, cyano and in each case straight-chain or branched alkyl or halogenoalkyl, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms, or in each case double-linked alkanediyl or alkenediyl, in each case having up to 4 carbon atoms; in addition, R^3 and R^4 independently of one another represent heterocyclylalkyl which has 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and 1 to 9 carbon atoms as well as 1 to 3 hetero atoms - in particular, nitrogen, oxygen and/or sulfur - in the heterocyclyl moiety and each of which is optionally monosubstituted or polysubstituted in the heterocyclyl moiety by identical or different substituents, whereby in each case suitable aryl substituents are: halogen, cyano, nitro, and in each case, straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy, halogenoalkylthio or alkoxycarbonyl, in each case having 1 to 5 carbon atoms and optionally 1 to 9 identical or different halogen atoms, and in addition R^3 and R^4 independently of one another represent aralkyl, aroyl or aryl, each of which has 6 to 10 carbon atoms in the aryl moiety and optionally 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted, identically or differently, whereby in each case suitable aryl substituents are: halogen, cyano, nitro, hydroxy, in each case straight-chain or branched alkyl, alkoxy, alkylthio, halogenoalkyl, halogenoalkoxy, halogenoalkylthio, alkylsulfinyl, alkylsulfonyl, halogenoalkylsulfinyl, halogenoalkylsulfonyl, alkanoyl or alkoxycarbonyl, in each case having 1 to 6 carbon atoms and optionally 1 to 9 identical or different halogen atoms, cycloalkyl having 3 to 6 carbon atoms or phenoxy, and whereby suitable alkyl substituents are: halogen or cyano, or

R^3 and R^4 together with the nitrogen atom to which they are bonded represent a five- to ten-membered heterocycle which can optionally contain 1 or 2 further hetero atoms, in particular, nitrogen, oxygen and/or sulfur, and which is optionally monosubstituted or polysubstituted, identically or differently, whereby suitable substituents are: halogen, and in each case straight-chain or branched alkyl or halogenoalkyl, in each case having 1 to 4 carbon atoms and optionally 1 to 9 identical or different halogen atoms as well as 1 to 2 oxo or thiono groups,

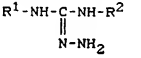
whereby the compounds in which

1. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, R^4 = cyclohexyl;

2. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, $R^4 = CH_2\text{-}C(CH_3)_3$;
3. $R^1 = CH_3$, $R^2 = C_2H_5$, $R^3 = H$, R^4 = 1-phenylethyl;
4. $R^1 = C_2H_5$, $R^2 = CH_3$, $R^3 = H$, $R^4 = C(CH_3)_3$;
5. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = CH_3$, $R^4 = C(CH_3)_3$;
6. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$, $R^4 = \text{-}(CH(CH_3)\text{-}CH = N\text{-}OCH_3$ and
7. $R^1 = CH_3$, $R^2 = CH_3$, $R^3 = H$,

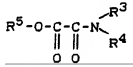
are excepted,
characterized in that

(a) amino guanidines of the general formula (II)



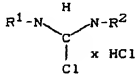
(II)

in which
R^1 and R^2 have the above-mentioned meaning,
and/or tautomers of the compounds of formula (II) and/or acid adducts of compounds of formula (II) or of tautomers thereof,
are reacted with oxalic acid ester amides of the general formula (III)



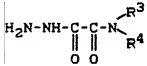
(III)

in which
R^3 and R^4 have the above-mentioned meanings and
R^5 represents alkyl,
optionally in the presence of a diluent and optionally in the presence of a reaction auxiliary, or when
(b) chloroformamidine hydrochlorides of the general formula (IV)



(IV)

in which
R^1 and R^2 have the above-mentioned meanings,
are reacted with oxalic acid amide hydrazides of the general formula (V)

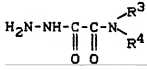


(V)

in which
R^3 and R^4 have the above-mentioned meanings,
optionally in the presence of a diluent and optionally in the presence of an acid acceptor, or when
(c) carbodiimides of the general formula (VI)

$$R^1\text{-}N = C = N\text{-}R^2 \qquad \text{(VI)}$$

in which
R^1 and R^2 have the above-mentioned meanings,
are reacted with oxalic acid amide hydrazides of the general formula (V)



(V)

in which
R^3 and R^4 have the above-mentioned meanings,
optionally in the presence of a diluent and optionally in the presence of a reaction auxiliary.

5. Herbicidal agent, characterized by a content of at least one 3-amino-5-aminocarbonyl-1,2,4-triazole derivative of formula (I) according to claim 1 or 4.

6. Method for the control of unwanted plants, characterized in that 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of formula (I) according to claim 1 or 4 is allowed to act upon the plants and/or their habitat.

7. Use of 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of formula (I) according to claim 1 or 4 for control of unwanted plants.

8. Method for the preparation of herbicidal agents, characterized in that 3-amino-5-aminocarbonyl-1,2,4-triazole derivatives of formula (I) according to claim 1 or 4 are mixed with extenders and/or surface-active agents.

European Patent Office

EUROPEAN SEARCH REPORT

Number of the Application

EP 90114303.2

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to Claim	CLASSIFICATION OF APPLICATION (Int. Cl.5)
X,P, D	EP - A1 - 0 332 991 (BAYER) * Claims 1,6-8 * --	1,5-7	C 07 D 249/14 C 07 D 401/06 C 07 D 405/06 C 07 D 409/06
A	EP - A1 - 0 048 555 (GLAXO) * Claim 1 * --	1	C 07 D 413/06 C 07 D 417/06 A 01 N 43/653
A	CHEMICAL ABSTRACTS, Band 85, No. 11, 13. September 1976, Columbus, Ohio, USA VORONKOV, M.G. et al. "Basicity and protonation center of 5(3)-substituted-3(5)-amino-1,2,4-triazole" page 487, column; 1, Abstract No. 77 387u & Dokl. Akad. Nauk SSSR 1976, 227(5), 1116-19 --	1	
A	CHEMICAL ABSTRACTS, Volume 76, No. 11, 13. March 1972, Columbus, Ohio, USA NICHOLSON, S. et al. "Covalent hydrates as transient species in heterocyclic rearrangements. I. Ring fission of s-triazolo-pyrazines" Page 381, column; 2, Abstract No. 58 494z & J. Chem. Soc., Perkin Trans 2 1972, (1), 4-11 ----	1	

FIELDS OF SEARCH (Int. Cl.5)

C 07 D 249/00
C 07 D 401/00
C 07 D 405/00
C 07 D 409/00
C 07 D 413/00
C 07 D 417/00

The present search report has been drawn up for all patent claims.

Place of search	Date of completion of the search	Examiner
VIENNA	September 28, 1990	HAMMER

CATEGORY OF CITED DOCUMENTS

- X: particularly relevant if taken alone
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- A: technological background
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